

# Direct measurement of non-linear properties of bipartite quantum states

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Non-linear properties of quantum states, such as entropy or entanglement, quantify important physical resources and are frequently used in quantum information science. They are usually calculated from a full description of a quantum state, even though they depend only on a small number of parameters that specify the state. Here we extract a non-local and a non-linear quantity, namely the Renyi entropy, from local measurements on two pairs of polarization entangled photons. We also introduce a “phase marking” technique which allows to select uncorrupted outcomes even with non-deterministic sources of entangled photons. We use our experimental data to demonstrate the violation of entropic inequalities. They are examples of a non-linear entanglement witnesses and their power exceeds all linear tests for quantum entanglement based on all possible Bell-CHSH inequalities.

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Many interesting properties of composite quantum systems, such as entanglement or entropy, are not measured directly but are inferred, usually from a full specification of a quantum state represented by a density operator. However, it is interesting to note that some of these properties can be measured in the same way we measure and estimate average values of observables. Here we illustrate this by extracting a non-local quantity, the Renyi entropy of the composite system, from local measurements on two pairs of polarization entangled photons. This quantity is a non-linear function of the density operator. We then use our experimental data to demonstrate the violation of entropic inequalities, which can be also interpreted as the experimental demonstration of a non-linear entanglement witness.

Consider a source which generates pairs of photons. The photons in each pair fly apart from each other to two distant locations A and B. Let us assume that the polarization of each pair is described by some density operator  $\varrho$ , which is unknown to us. Following Schrödinger’s remarks on relations between the information content of the total system and its sub-systems [1], it has been proven that for separable states global von Neumann entropy is always not less than local ones [2]. Subsequently a number of entropic inequalities have been derived, satisfied by all separable states [3, 4, 5, 6]. The simplest one is based on the Renyi entropy, or the purity measure,  $\text{Tr}(\varrho^2)$  and can be rewritten as

$$\text{Tr}(\varrho_A^2) \geq \text{Tr}(\varrho^2), \text{Tr}(\varrho_B^2) \geq \text{Tr}(\varrho^2), \quad (1)$$

where  $\varrho_A$  and  $\varrho_B$  are the reduced density operators pertaining to individual photons. The inequalities (1) involve non-linear functions of density operators and are known to be stronger than *all* Bell-CHSH inequalities [3, 7]. There are entangled states which are not

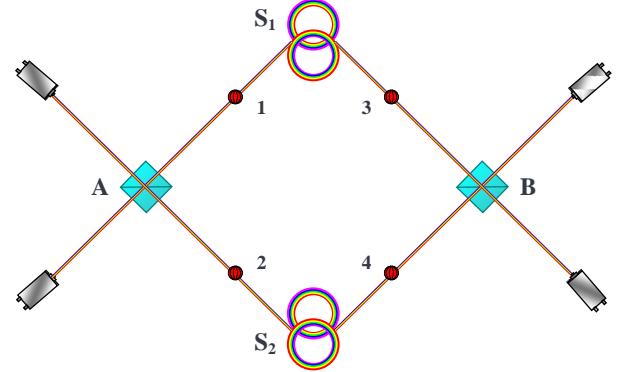


FIG. 1: An outline of our experimental set-up. Sources  $S_1$  and  $S_2$  emit pairs of polarization-entangled photons. The entangled pairs are emitted into spatial modes 1 and 3, and 2 and 4. One photon from each pair is directed into location A and the other into location B. At the two locations photons impinge on beam-splitters and are then detected by photodetectors. There are four possible outcomes in this experiment: coalescence at A and coalescence at B, coalescence at A and anti-coalescence at B, anti-coalescence at A and coalescence at B, anti-coalescence at A and anti-coalescence at B. The probabilities associated with the four outcomes are, respectively,  $p_{cc}$ ,  $p_{ca}$ ,  $p_{ac}$ ,  $p_{aa}$  (subscript  $b$  stands for coalescence and  $a$  for anticoalescence). In terms of these probabilities the entropic inequalities (1) are written as  $p_{ca} \geq p_{aa}$ ,  $p_{ac} \geq p_{aa}$ .

detected by the Bell-CHSH inequalities but which are detected by the entropic inequalities (1). [14]

In the experiment a source generates pairs of polarization-entangled photons in a singlet state  $|H\rangle|V\rangle - |V\rangle|H\rangle$ , where  $H$  and  $V$  stand for horizontal and vertical polarizations respectively. Thus  $\rho$  is a maximally entangled pure state and, consequently,  $\rho_A = \rho_B$

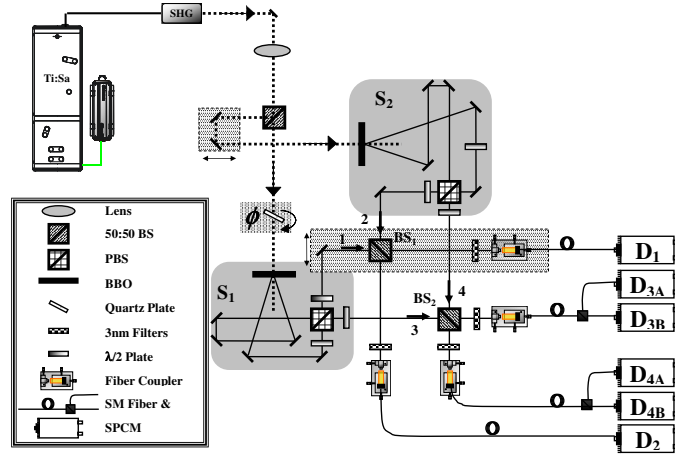


FIG. 2: Experimental set-up. Two pairs of polarization entangled photons are generated in two separate 3mm thick BBO crystals via typeII spontaneous parametric down conversion (SPDC). The crystals are pumped by two beams, each consisting of a train of femtosecond laser pulses with the central wavelength of 415nm and a repetition rate of 76MHz (12.5ns between pulses). The beams are displaced by few micrometers using a trombone and the sub wavelength path difference (phase shift  $\phi$ ) is introduced by a tilted 1mm thick quartz-plate inserted into one of the pump beams. The two pump beams are obtained by splitting the main beam (average power 470mW), which is generated by doubling the frequency of the output of a Coherent MIRA-VERDI laser (operating in the mode-locking regime, 830nm central wavelength, 9nm bandwidth, and 160fs of pulse duration) using second harmonic generation (SHG) in a 2mm-thick LBO crystal. Entangled photons from the BBO crystals enter interferometers which provide high fidelity polarization entangled states in ultra-fast regime [12]. Subsequently the photons are sent to the beam-splitters  $BS_1$  and  $BS_2$ , so that each beam-splitter receives two photons, one photon from each entangled pair. The photons are coupled in Single-Mode (SM) Fibers [13] and detected by the silicon-based single-photon counting modules (Perkin-Elmer SPCM-AQR-14). Once the correct position of the two beam splitters is established the four-fold coincidence interference patterns are measured. Coincidence counts  $D_1 \otimes D_2 \otimes D_{3A} \otimes D_{4A}$ ,  $D_1 \otimes D_2 \otimes D_{3A} \otimes D_{4B}$ ,  $D_1 \otimes D_2 \otimes D_{3B} \otimes D_{4A}$  and  $D_1 \otimes D_2 \otimes D_{3B} \otimes D_{4B}$  allow to estimate the rate of anticoincidence-anticoincidence events, whereas  $D_1 \otimes D_2 \otimes D_{3A} \otimes D_{3B}$  and  $D_1 \otimes D_2 \otimes D_{4A} \otimes D_{4B}$  give the anticoincidence-coalescence rate.

are maximally mixed, completely depolarized states. In order to measure a quadratic property of  $\rho$  we need to operate on at least two copies of the entangled pairs. Here we use the phenomenon of coalescence and anti-coalescence of photons. If two identical photons are incident on two different input ports of a beam-splitter they will coalesce i.e. they will emerge together in one of the two, randomly chosen, output ports. More precisely, all pairs of photons (in general all pairs of bosons) with a symmetric (under the exchange of photons) polarization state will coalesce and all pairs of photons with an anti-symmetric polarization state will anti-coalesce i.e. photons will emerge separately in two different output ports of the beam-splitter. A beautiful experimental observation of this effect was reported by Hong, Ou, and Mandel over fifteen years ago [8], and more recently by Di Giuseppe et al [9]. The main idea behind our experiment is illustrated in Fig. (1). Two independent pairs of photons are generated by sources  $S_1$  and  $S_2$ , and one photon from each pair is directed into location A and the other into location B. At the two locations the photons impinge on beam-splitters and are then detected by photo-detectors. The beam-splitters at A and B, as long as the photons from two different pairs arrive within the coherence time, effectively project on the symmetric and anti-symmetric subspace in the four dimensional Hilbert

space associated with the polarization degrees of freedom. Let us consider the four possible detections in this experiment: coalescence at A and coalescence at B, coalescence at A and anti-coalescence at B, anti-coalescence at A and coalescence at B, and finally, anti-coalescence at A and anti-coalescence at B. Let the probabilities associated with the four outcomes be, respectively  $p_{cc}$ ,  $p_{ca}$ ,  $p_{ac}$ ,  $p_{aa}$ , and (subscript  $c$  stands for coalescence and  $a$  for anti-coalescence). In technical terms they correspond to probabilities of projecting the state  $\rho \otimes \rho$  of two pairs of photons on symmetric or antisymmetric subspaces at location A (associated with spatial modes 1 and 2) and B (associated with spatial modes 3 and 4), e.g.  $p_{ca} = \text{Tr}(P_S \otimes P_A)(\rho \otimes \rho)$  etc, where  $P_S$  and  $P_A$  are the corresponding projectors on the symmetric and antisymmetric subspaces. We can now write

$$\text{Tr } \rho^2 = p_{cc} - p_{ca} - p_{ac} + p_{aa}, \quad (2)$$

$$\text{Tr } \rho_A^2 = p_{cc} + p_{ca} - p_{ac} - p_{aa}, \quad (3)$$

$$\text{Tr } \rho_B^2 = p_{cc} - p_{ca} + p_{ac} - p_{aa}, \quad (4)$$

and the inequalities (1) can be expressed in a new and simple form,

$$p_{ca} \geq p_{aa} \quad , \quad p_{ac} \geq p_{aa} \quad (5)$$

Theoretical predictions for the singlet state are  $p_{cc} = 3/4$ ,

$p_{ac} = p_{ca} = 0$  and  $p_{aa} = 1/4$ . Let us mention in passing that in this particular case a coincidence count at A (B) projects the state of the remaining two photons at B (A) on the singlet state, inducing an entanglement swapping, c.f. [10]. The polarization entangled photons are generated using type-II spontaneous parametric down conversion (SPDC) [11]. An ultraviolet pulse from a pump laser is split into two pulses which are slightly delayed with respect to each other and directed towards two  $\beta$ -barium-borate (BBO) crystals. The two BBO crystals correspond to the two sources  $S_1$  and  $S_2$ . When the pulses pass through the crystals they emit, with some probability, pairs of energy-degenerate polarization-entangled photons into modes 1 and 3 (source  $S_1$ ), and 2 and 4 (source  $S_2$ ). Modes 1 and 2 are coupled by the beam-splitter at A and modes 3 and 4 by the beam-splitter at B. Behind the beam-splitters silicon single-photon counting modules register emerging photons. The coalescence and anti-coalescence coincidences are recorded. (See Fig. (2) for technical details.) Currently available sources of entangled photons are probabilistic which creates an additional experimental difficulty. When a UV pulse passes through a BBO crystal it produces a superposition of vacuum, two entangled photons, four entangled photons, etc. A four-photon coincidence in our set-up may be caused by two entangled pairs from two different sources but also by four photons from one source and no photons from the other, as shown in Fig. (3), moreover, the three scenarios are equally likely. In order to discriminate unwelcome four-photon coincidences we have used “phase marking” - for certain values of the phase difference between the two pumping beams we register only coincidences that not corrupted by the spurious emissions. The description can be made more quantitative by analysing an effective Hamiltonian describing entanglement generation in two coherently pumped BBO crystals,

$$H = \eta(K + K^\dagger) + \eta(Le^{-i\phi} + L^\dagger e^{i\phi}). \quad (6)$$

Here  $\eta$  is a coupling constant, proportional to the amplitude of the pumping beams,  $\phi$  is the relative phase shift between the beams introduced by the tilted quartz-plate, and  $K = a_{1H}a_{3V} - a_{1V}a_{3H}$  and  $L = a_{2H}a_{4V} - a_{2V}a_{4H}$  are the linear combination of annihilation operators describing the down-converted modes. The subscripts 1, 2, 3, 4 label the spatial modes and  $H, V$  stand for horizontal and vertical polarizations. The four-photon term of a quantum state generated by this Hamiltonian can be written as

$$\begin{aligned} |\Psi\rangle = & \frac{e^{i\phi}}{\sqrt{10}}(a_{1H}^\dagger a_{3V}^\dagger - a_{1V}^\dagger a_{3H}^\dagger)(a_{2H}^\dagger a_{4V}^\dagger - a_{2V}^\dagger a_{4H}^\dagger) \\ & + \frac{1}{\sqrt{10}} \left( \frac{1}{2} a_{1H}^{\dagger 2} a_{3V}^{\dagger 2} - a_{1H}^\dagger a_{1V}^\dagger a_{3V}^\dagger a_{3H}^\dagger + \frac{1}{2} a_{1V}^{\dagger 2} a_{3H}^{\dagger 2} \right) \\ & + \frac{e^{2i\phi}}{\sqrt{10}} \left( \frac{1}{2} a_{2H}^{\dagger 2} a_{4V}^{\dagger 2} - a_{2H}^\dagger a_{2V}^\dagger a_{4V}^\dagger a_{4H}^\dagger + \frac{1}{2} a_{2V}^{\dagger 2} a_{4H}^{\dagger 2} \right) \\ & |vac\rangle, \end{aligned} \quad (7)$$

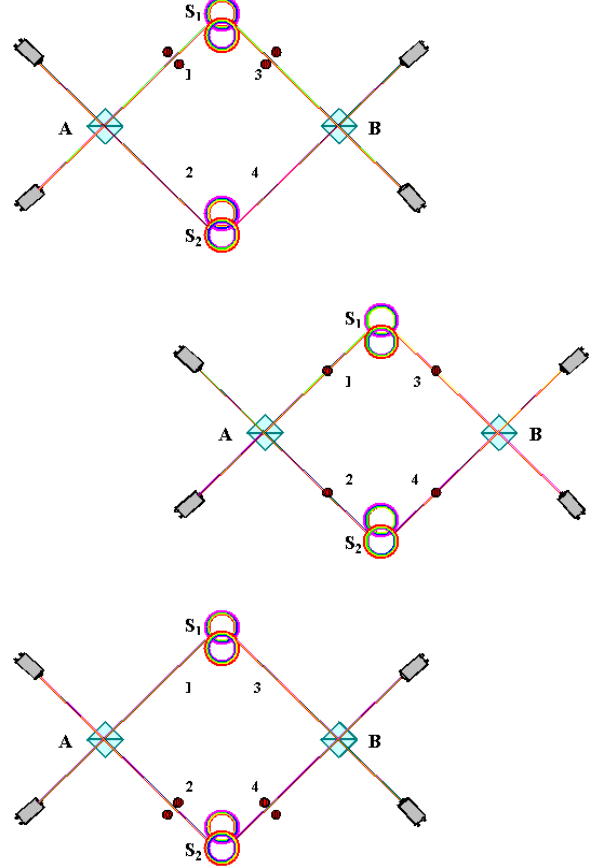


FIG. 3: Possible emissions leading to four-photons coincidences. Parametric down-conversion is not an ideal source of entangled photons. It generates a superposition of vacuum, two-entangled photons, four-entangled photons, etc. The central diagram shows the desired emission of two independent entangled pairs – one by source  $S_1$  and one by source  $S_2$ . The top and the bottom diagrams show unwelcome emissions of four photons by one of the two sources. Although the three cases are equally likely we can eliminate contributions of unwelcome emissions by a judicious choice of the phase difference between the beams that coherently pump the two BBO crystals. We call it “phase marking”. Symmetric superposition of the unwelcome emissions, obtained for  $\phi = 0$ , does not contribute to the coalescence-anticoalescence events, whereas the antisymmetric superposition, obtained for  $\phi = \pi/2$  does not affect the anticoalescence-anticoalescence outcomes.

where the first term describes the desired two polarization-entangled pairs, each in the singlet state  $|H\rangle|V\rangle - |V\rangle|H\rangle$ , whereas the last two terms describe unwelcome four-photon states generated by an emission from only one of the two crystals (see Fig.3). The coalescence and anti-coalescence coincidences for the state (7) are given by

$$p_{ac} = p_{ca} = \frac{3}{20}(1 - \cos 2\phi), p_{aa} = \frac{1}{4} + \frac{3}{20} \cos 2\phi. \quad (8)$$

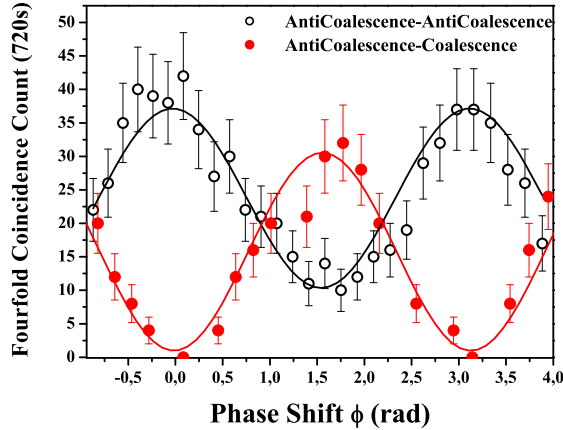


FIG. 4: Four-photon coincidences. The plot shows the four-photon interferences as a function of the pump displacement which is proportional to the relative phase shift  $\phi$  of the two pumps. The probability of coalescence and anti-coalescence for ideal singlet state case can be extracted from the coincidence counts at the minima of interference curves using (8). The minima of the curves correspond to the required values of  $p_{ac}$  and  $p_{aa}$ . The experimental data show that  $p_{aa} > p_{ac}$ , which violates the entropic inequalities (1). The results for the curve anticoalescence-coalescence are normalized by a factor 2 to take into account the fact that half of the coincidences are lost.

In order to recover the coincidences associated with the desired singlet state we notice that for  $\phi = 0$  and  $\phi = \pi/2$  there are no spurious contributions to  $p_{ac} = p_{ca}$  and  $p_{aa}$  respectively. For these two phase settings the symmetric and antisymmetric superposition of the last two terms in (7) lead to additional symmetries at the input of the beam-splitters and cancels out the unwelcome outcomes.

In the experiment we have traced the dependence of  $p_{ac}(\phi)$  and  $p_{aa}(\phi)$ . Even though the measurement of  $p_{ac} = p_{ca}$  and  $p_{aa}$  could only be realized for different values of the phase  $\phi$ , the difference in probability values is clearly observed by following the minima of both curves in Fig. (4). Based on the statistical fit of curves in Fig. (4) we obtain  $p_{ac} = p_{ca} = 0.0255 \pm 0.008$  and  $p_{aa} = 0.2585 \pm 0.008$ . The measured result is in good agreement with the theoretical prediction and clearly demonstrates the violation of the entropic inequalities for the singlet state. Let us stress that these inequalities involve nonlinear functions of a quantum state. Their power exceeds all linear tests such as the Bell-CHSH inequalities with all possible settings and entanglement witnesses. In fact our result can be viewed as the first experimental measurement of a non-linear entanglement witness. Direct measurements of non-linear properties of quantum states open new ways of probing and manipulating quantum phenomena.

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  - [14] For example, if we mix the maximally entangled singlet state with the maximally depolarized state, in proportions  $p$  and  $1 - p$  respectively ( $0 \leq p \leq 1$ ), then the resulting state is entangled for  $p > \frac{1}{3}$ . However, the entanglement is detected by the CHSH inequalities for  $p > \frac{1}{\sqrt{2}} \approx 0.707107$  and by the non-linear inequalities for  $p > \frac{1}{\sqrt{3}} \approx 0.57735$ .